

THE CASIMIR EFFECT AND THE FOUNDATIONS OF STATISTICAL PHYSICS

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The Lifshitz theory and its modifications are discussed with respect to the Nernst heat theorem and the experimental data of several recent experiments. An analysis of all available information leads to the conclusion that some concepts of statistical physics might need reconsideration.

Keywords: Lifshitz theory; Casimir force; Nernst heat theorem.

1. Introduction

In the last few years there has been an explosion of interest in the Casimir effect¹ which has resulted in new precise experiments, elaboration of powerful theoretical methods and in suggestions of prospective applications (a modern overview of the subject can be found in Ref. 2). Coincident with many developments of a conclusive character, starting from 2000 there were also controversial discussions in the literature on the nature and size of the thermal effects in the Lifshitz theory of the Casimir force.^{2,3} Boström and Sernelius⁴ were the first who predicted the existence of large thermal corrections to the Casimir force between two plane parallel metallic plates described by the Drude model spaced at separation of a few hundred nanometers. Bordag et al.⁵ argued that such corrections are nonphysical and suggested to calculate the thermal Casimir force using the dielectric permittivity of the plasma model (for the latter purpose the plasma model was also used in Ref. 6). Later both approaches were further developed in Refs. 7, 8 and 9, 10, respectively. A step of paramount importance was made by the experiments of Decca et al.^{11,12} which excluded the existence of large thermal corrections predicted by the Drude model at almost 100% confidence level. Related experiments for semiconductor¹³ and dielectric¹⁴ materials leading to similar conclusions¹⁵ were subsequently performed. On the one hand, thermodynamic arguments based on the Nernst heat theorem favored the plasma model approach for metals¹⁰ and neglect of the dc conductivity for dielectrics.¹⁶ On the other hand, statistical physics applied in the so-called classical limit was in support of the Drude model.¹⁷ The situation was so extraordinary that it was even suggested^{18–20} to modify the Lifshitz theory pro-

viding the fundamental description of both the van der Waals and Casimir forces between real materials. For this purpose the standard reflection coefficients were replaced with their generalizations taking into account the screening effects and diffusion currents. It was shown, however, that the modified theory still violates the Nernst heat theorem^{21–26} and is in contradiction with the experimental data. These conclusions were disputed^{27–29} by the authors of the modified theory.

Keeping in mind that controversial discussion on this subject has lasted for already ten years and consensus is not yet achieved, it seems pertinent to collect and analyze all the proposed arguments. Such an analysis seems to be especially useful because there were discussions in the previous literature which appear one sided by dealing with only selected facts and disregarding others. By taking into account all known facts in a fair manner (i.e., by assuming that published experimental and theoretical results are correct if we cannot indicate any specific mistake invalidating them), we arrive at the conclusion that some of the concepts of statistical physics commonly used for the theoretical description of the interaction of fluctuating fields with matter need to be reconsidered.

The structure of this paper is as follows. In Sec. 2 we briefly discuss the Nernst theorem in the Lifshitz theory. Sec. 3 is devoted to the same subject in application to the proposed modifications of the Lifshitz theory. In Sec. 4 we consider what the experiments say and if they are reliable. Sec. 5 considers what statistical physics says. Sec. 6 contains our conclusions.

2. The Lifshitz theory and the Nernst heat theorem

The Lifshitz theory provides an expression for the free energy $\mathcal{F}(a, T)$ of the fluctuating electromagnetic field interacting with two thick uncharged plates (semispaces) separated by a gap of width a per unit area of plates. It is supposed that this system is in thermal equilibrium at temperature T . Material of the plates is described by the dielectric permittivity $\varepsilon(\omega)$ depending only on the frequency. Under these conditions $\mathcal{F}(a, T)$ is expressed in terms of the Fresnel reflection coefficients $r_{\text{TM,TE}}(i\xi_l, k_\perp)$ for the transverse magnetic (TM) and electric (TE) polarizations of the electromagnetic field calculated at the imaginary Matsubara frequencies $\xi_l = 2\pi k_B T l / \hbar$, where k_B is the Boltzmann constant, $l = 0, 1, 2, \dots$, and $\mathbf{k}_\perp = (k_x, k_y)$ is the projection of the wave vector on the plane of the plates.²

For materials with no free charge carriers (insulators) $\varepsilon(i\xi)$ can be represented in the oscillator form

$$\varepsilon(i\xi) = 1 + \sum_{j=1}^K \frac{g_j}{\omega_j^2 + \xi^2 + \gamma_j \xi}, \quad (1)$$

where $\omega_j \neq 0$ are the oscillator frequencies and $\varepsilon_0 \equiv \varepsilon(0) < \infty$. Electrons in metals are usually described by the Drude or plasma models

$$\varepsilon_D(i\xi) = 1 + \frac{\omega_p^2}{\xi(\xi + \gamma)}, \quad \varepsilon_p(i\xi) = 1 + \frac{\omega_p^2}{\xi^2}, \quad (2)$$

where ω_p is the plasma frequency, γ is the relaxation parameter.

It was suggested^{9,10} to use the Nernst heat theorem as a test of applicability of different models of ε in the Lifshitz theory. The entropy of the system under consideration (the two plates interacting with the fluctuating field) per unit area of plates is finite and can be calculated as

$$S_{\text{syst}}(a, T) = -\frac{\partial \mathcal{F}(a, T)}{\partial T} - \frac{\partial \mathcal{F}_n(T)}{\partial T} \equiv S(a, T) + S_n(T). \quad (3)$$

Here, $S(a, T)$ is the separation-dependent part of the entropy related to the interaction between the fluctuating field and the plates, and $\mathcal{F}_n(S_n)$ are the parts of the free energy (entropy) of the system which do not depend on a . The quantities $\mathcal{F}_n(S_n)$ are related to the noninteracting case (specifically, they contain the large free energy and entropy of remote plates) and do not contribute to the Casimir force.

There are different formulations of the third law of thermodynamics (the Nernst heat theorem) in the literature (some of them are discussed in Ref. [30]). Below throughout the text we use only the standard formulation from textbooks which is the following.^{31,32} When $T \rightarrow 0$, the entropy of an equilibrium system [in our case $AS_{\text{syst}}(a, T)$ where A is the area of the plates] goes to a finite limit $S_{\text{syst},0}$ which does not depend on volume, pressure, density or other thermodynamic parameters of the system. According to quantum statistical physics, we get^{31,32}

$$S_{\text{syst},0} = k_B \ln W_0, \quad (4)$$

where W_0 is an integer number describing the degree of degeneracy of the ground state of the system. If the ground state is nondegenerate, $W_0 = 1$, one has $S_{\text{syst},0} = 0$. The latter, however, is not necessary to satisfy the Nernst theorem as formulated above. It is important only that $S_{\text{syst},0}$ does not depend on the continuous thermodynamic parameters, specifically, on a . Keeping in mind that $S(a, T)$ is the part of the entropy depending on a , the necessary requirement for the satisfaction of the Nernst theorem is that $S(a, T) \rightarrow 0$ when $T \rightarrow 0$. If $S(a, T)$ goes to some function of a , $f(a)$, when $T \rightarrow 0$, the Nernst theorem is violated because $f(a)$ cannot be compensated by the a -independent limit of the quantity $S_n(T)$.

When ε_D of the Drude model (2) is substituted into the Lifshitz formula for metals with perfect crystal lattices, we get¹⁰

$$S(a, 0) = S_D(a, 0) = -\frac{k_B \zeta(3)}{16\pi a^2} \left[1 - 4\frac{\delta_0}{a} + 12\left(\frac{\delta_0}{a}\right)^2 - \dots \right] < 0, \quad (5)$$

where $\delta_0 = c/\omega_p$ is the skin depth and $\zeta(z)$ is the Riemann zeta function. A metal with perfect crystal lattice is a truly equilibrium system. Thus in this case we deal with the violation of the Nernst heat theorem. It was argued in the literature²⁷ that with the decrease of T the frequency region of the anomalous skin effect, where local description by means of $\varepsilon_D(\omega)$ is inapplicable, extends to low frequencies. This objection, however, does not solve the problem. First, for any low T there exists

some narrow region of small frequencies $[0, \omega_0]$ where local description by means of $\varepsilon_D(\omega)$ is applicable. Then the result (5) remains valid because it originates from the zero-frequency term of the Lifshitz formula. Second, the Drude model at low T was used for the interpolation between the regions of the normal skin effect and infrared optics in the classical theories by Bloch, Grüneisen and Debye.³³ Although such a model approach does not provide an exact description of real metals due to the existence of the anomalous skin effect, it seems strange that it leads to the violation of the Nernst theorem when used in combination with the Lifshitz formula. Note that for metals with impurities the Lifshitz formula combined with the Drude model satisfies the Nernst theorem.^{8,34} This is a step forward in the resolution of the problem but does not solve it because the introduction of impurities might result in a violation of the thermal equilibrium which for sure takes place for perfect crystal lattices (see a discussion³⁵). At the same time the substitution of ε_p of the plasma model (2) into the Lifshitz formula leads to $S_p(a, 0) = 0$. For insulators it was shown¹⁶ that $S(a, T)$ calculated with the dielectric permittivity (1) goes to zero when T vanishes. If, however, the dc conductivity $\sigma_0(T)$ is taken into account,

$$\varepsilon_{\text{dc}}(i\xi, T) = \varepsilon(i\xi) + 4\pi \frac{\sigma_0(T)}{\xi}, \quad (6)$$

it results in the violation of the Nernst theorem

$$S(a, 0) = S_{\text{dc}}(a, 0) = \frac{k_B}{16\pi a^2} [\zeta(3) - \text{Li}_3(r_0^2)] > 0. \quad (7)$$

Here, $\text{Li}_3(z)$ is the polylogarithm function and $r_0 = (\varepsilon_0 - 1)/(\varepsilon_0 + 1)$.

To avoid the violation of the Nernst theorem and contradictions with the experimental data (see Sec. 4) in numerous applications of the Lifshitz theory, the following phenomenological prescription was proposed.^{36,37} When applying the Lifshitz theory to metals, conduction electrons should be described by the plasma model. In the application of this theory to dielectrics, dc conductivity should be omitted. Keeping in mind that all materials can be divided into metals (whose conductivity is not equal to zero at $T = 0$) and dielectrics (whose conductivity vanishes when $T \rightarrow 0$), this prescription can be considered as universally applicable. In some sense it is not new because metals were often described in the literature by means of the plasma model^{6,38} and the dc conductivity of dielectrics was almost always omitted.¹⁴ It was generally believed, however, that with account of relaxation properties of conduction electrons (i.e., using the Drude model) and of the dc conductivity of dielectrics slightly more exact results would be obtained. The new fact recognized in the last few years is that the inclusion of these features leads to drastically different calculational results which are in conflict with thermodynamics and contradict the experimental data. This fact invites reconsideration of the Lifshitz theory and careful analysis of all assumptions laid in its foundation.

3. The Nernst heat theorem in the modifications of the Lifshitz theory

The most general modification¹⁹ leaves the formalism of the Lifshitz theory unchanged but replaces the Fresnel reflection coefficients, $r_{\text{TM,TE}}(i\xi_l, k_\perp)$ with the modified ones, $\tilde{r}_{\text{TM,TE}}(i\xi_l, k_\perp)$, which take into account both the drift and diffusion currents by means of the Boltzmann transport equation. The modified reflection coefficients depend on a new parameter κ which has the physical meaning of an inverse screening radius. It is equal to κ_{DH} or κ_{TF} for Debye-Hückel and Thomas-Fermi screening radii applicable for the Maxwell-Boltzmann and Fermi-Dirac statistics, respectively. For dielectrics ($\kappa = \kappa_{\text{DH}}$) at $\xi = 0$ the coefficient $\tilde{r}_{\text{TM}}(0, k_\perp)$ was first obtained in Ref. 18. The modified reflection coefficients $\tilde{r}_{\text{TM,TE}}$ were also phenomenologically expressed²⁰ in terms of \mathbf{k} -dependent dielectric permittivities in the random phase approximation (recall that in the presence of a gap between semispaces the translation invariance in space is violated and the nonlocal dielectric permittivity ε_z depending on \mathbf{k} does not exist as a rigorous mathematical concept³⁹).

For metals with perfect crystal lattice by using $\kappa = \kappa_{\text{TF}}$ it was shown^{22,23} that the modified entropy $\tilde{S}(a, 0) = S_D(a, 0) < 0$, as can be seen from Eq. (5). Thus, in this case the modification of the Lifshitz theory proposed^{18–20} suffers from the same thermodynamic difficulty as the standard Drude model. For dielectric materials ($\kappa = \kappa_{\text{DH}}$) the situation turned out to be more involved. Under the condition that the density of charge carriers $n(T) \rightarrow 0$ more quickly than $T^{1+\alpha}$ with $\alpha > 0$ (this is the case for intrinsic semiconductors) it was shown^{21,24} that the modified entropy $\tilde{S}(a, 0) = 0$, i.e., the Nernst theorem is satisfied. In the two Comments^{25,26} it was stressed, however, that for dielectric materials not satisfying this condition (for instance, for doped semiconductors with $n < n_{\text{cr}}$, semimetals of dielectric type and solids with ionic conductivity) the modified Lifshitz theory violates the Nernst heat theorem. In this case it holds $\tilde{S}(a, 0) = S_{dc}(a, 0) > 0$ where S_{dc} is defined in Eq. (7). For dielectric materials under consideration $n(T)$ does not go to zero with vanishing T and conductivity vanishes with temperature due to the vanishing mobility of charge carriers.

The result that the modifications of the Lifshitz theory are in disagreement with thermodynamics was disputed in the literature. Thus, it was claimed²⁹ that the approach of Ref. 19 satisfies the Nernst theorem for all dielectrics. However, in the respective proof it was assumed that $n(T) \rightarrow 0$ when $T \rightarrow 0$. The above-mentioned dielectric materials for which this is not the case were not discussed. Reply²⁷ claimed that the materials leading to conflicts with thermodynamics in the modified Lifshitz theory are amorphous glass-like bodies which are out of equilibrium state and have a big entropy at $T = 0$. The Nernst theorem is not valid for such bodies. It is true that glass-like bodies must not satisfy the Nernst theorem. The arguments in the Reply²⁷ are, however, somewhat contradictory. The point is that we consider not the entropy $S_n(T)$ of the plate made of a glass-like material

(SiO₂ for instance¹⁴), but the entropy of the interaction with the fluctuating field $S(a, T)$ (see Sec. 2). If the fluctuating field is in equilibrium with the plate (as is assumed in Ref. 14), one can apply the Lifshitz theory. In this case, however, in accordance with the Nernst theorem, $S(a, T)$ must vanish when T vanishes. In fact the input data for the Lifshitz formula are the values of $\varepsilon(i\xi)$ which are quite similar for the amorphous and polycrystal SiO₂. The Lifshitz formula is applicable when the fluctuating field is in equilibrium with the material of the plate. This formula is incapable of distinguishing between the cases when the plate material is in equilibrium or out of equilibrium. Reply²⁷ does not also provide a response concerning the existence of crystalline materials (semimetals of the dielectric type, for instance) leading to the violation of the Nernst theorem in the proposed modifications of the Lifshitz theory.

Both Replies^{27,28} cast doubts on the fact that there are dielectric materials for which $n(T)$ does not go to zero when $T \rightarrow 0$ with a reference to the measurements⁴⁰ for SiO₂ performed in the region from 433 K to 473 K. Such high- T results seem to be irrelevant to the problem under consideration. Independent measurements of all three parameters, conductivity, n and mobility, demonstrate⁴¹ that “mobility has the dominant influence upon the conductivity-temperature dependence.” As was recently confirmed,⁴² “On long time scales the ‘mobile’ ion density must be the total ion concentration. This ‘long run’ may be years or more, and ions trapped for so long are for all practical purposes immobile. Nevertheless, unless there are infinite barriers in the solid, which is unphysical, in the very long run all ions are equivalent.” Thus, for ionic conductors (like amorphous SiO₂) n does not vanish when $T \rightarrow 0$. The same conclusion holds for compensated semiconductors of the dielectric type. If the density of donor atoms n_d is larger than the density of acceptor atoms n_a , the density of charge carriers at low T , $n_d - n_a$, remains constant.⁴³ One more example is provided by semimetals of the dielectric type which are crystal materials with a regular structure. For these materials the Fermi energy is at a band where the density of states is not equal to zero. The number of charge carriers near the Fermi surface is fixed and determined by the structure of the crystal lattice. For both compensated semiconductors and semimetals of dielectric type conductivity vanishes due to vanishing mobility.^{44,45} All the above testifies that the problem of thermodynamic inconsistency of the proposed modifications of the Lifshitz theory deserves serious attention.

4. What experiments say and is it reliable

It was widely discussed in the literature that the measurement data of the experiments with a micromechanical oscillator^{11,12} exclude the use of the Drude model for the calculation of the thermal Casimir force between metals but are consistent with the use of the plasma model. The experiments with an atomic force microscope¹³ and Bose-Einstein condensate¹⁴ are inconsistent with the inclusion of the dc conductivity of a dielectric plate but consistent with the theory omitting this conductivity.

These results are related to the standard Lifshitz theory. They are obtained at a 99.9% and 95% confidence levels with respect to experiments of Refs. 12, 13 and at a 70% confidence level for the experiment of Ref. 14.

Just after the modifications of the Lifshitz theory were proposed, the obtained theoretical results (which are almost coincident for all three variants of the modified theory) were compared with the experimental data. For metals, it was found^{22,23,26} that the experimental data¹² exclude the modified Lifshitz theory at a 99.9% confidence level. For dielectrics, the data of the experiment¹³ exclude the predictions of the modified theory at a 70% confidence level.^{21,24,25} It was found also that the data of the experiment¹⁴ determined at a 70% confidence level are not precise enough and do not permit to make a conclusive comparison with theory. The point is that it is consistent with both the standard Lifshitz theory with dc conductivity excluded and with the modified Lifshitz theory.

Note that it was claimed²⁰ that the experimental data¹³ can hardly distinguish between the standard Lifshitz theory with omitted dc conductivity of dielectric Si and the modified theory. This claim is based on a complete misunderstanding of statistical procedures used for the comparison between experiment and theory. Thus, in Fig. 1a of Ref. 20 the experimental data are shown with errors determined at a 70% confidence level, but the width of the theoretical band related to the modified theory was calculated at a 95% confidence level (i.e., artificially widened in order to make theory consistent with the data). Such a comparison is evidently irregular. In the Erratum,²⁰ instead of plotting the theoretical band at a 70% confidence level, the experimental errors were increased by calculating them at a 95% confidence level. This is, however, meaningless because the data¹³ are not of sufficient precision for the conclusive comparison with the modified Lifshitz theory at a 95% confidence level.²¹ If a comparison at the 70% confidence level would be made, the result²¹ on the exclusion of the modified theory is reproduced.

Thus, the Drude model approach and the modified Lifshitz theory are in disagreement with the experimental data. The question arises what is the reliability of these experiments. The experiments under consideration were repeated several times with the same result and the most conservative statistical procedures for the data processing and error analysis have been used. It was claimed, however, that there is an anomalous distance dependence of the gradient of the electric force, used for calibration of the Casimir setup, between an Au plate and an Au spherical lens of 30 mm radius.⁴⁶ The respective contact potential was found to be separation-dependent. On this basis it was suggested to perform a reanalysis of the previous experiments mentioned above. These doubts cast on previous experiments with small spheres of about 100 μm radius are not justified. The reason is that the contact potential in the experiments^{11,12} was measured to be constant over a wide range of separations and the standard force-distance dependence for the electric force was observed, as predicted by classical electrodynamics. The possible reason for the anomalous dependence observed⁴⁶ is deviation of the mechanically polished and ground surface of the centimeter-size radius from a perfect spherical shape.⁴⁷

An attempt to avoid this conclusion using the capacitance measurements at large separations⁴⁸ was shown to be based on incorrect computations.⁴⁹ Because of this, continuing claims that important systematic effects have not been properly taken care of in the electrostatic calibrations in previous experiments, in our opinion, are unfair and cannot be considered as a scientific argument against these experiments. This does not mean that there is no need to look for systematic effects which might be present in previously performed experiments. It would be desirable, however, that such kind investigations were performed in the experimental configurations maximally similar to the original ones and were not based on far-reaching extrapolations.

5. What statistical physics says

Classical statistical physics permits one to calculate the free energy for two remote plates consisting of mobile quantum charges interacting with the quantized electromagnetic field. In doing so, photons and charges are supposed to be in thermal equilibrium at temperature T . The obtained free energy¹⁷ is equal to the one calculated by using the Lifshitz formula combined with the Drude model (i.e., equal to one half of the result valid for ideal metal plates).

Another consequence of statistical physics is the Bohr-van Leeuwen theorem which states that in classical systems at thermal equilibrium matter decouples from the transverse electromagnetic field. Recently it was shown⁵⁰ that this theorem is satisfied if and only if at large separations the reflection coefficient $r_{\text{TE}}(0, k_{\perp})$ of nonmagnetic materials is equal to zero leading to the same result for the free energy as the Lifshitz formula combined with the Drude model. Thus, in the classical limit (at large separations) the Drude model approach finds support from the source side of statistical physics although it has difficulties with respect to the Nernst theorem and disagrees with the experimental data at short separations.

It is useful to reformulate the problem in an equivalent way. It was shown⁵¹ that large negative temperature correction arising in the Drude model approach at short separations can be described as the contribution of eddy currents. The absence of this contribution in the measurement data was interpreted in a way that it was somehow reduced.⁵¹ The mechanism of this reduction remains, however, unclear. As a possible resolution of the problem the standard Planck distribution was modified⁵² by including a phenomenological parameter D taking into account the “saturation effects”. In this way an agreement between the Lifshitz theory combined with the Drude model and experimental data^{12–14} was achieved. However, the relative arbitrariness in the value of D remains a problem.

The roots of the controversial situation under consideration might be connected with the use of some basic statements of statistical physics outside of their application region. It is common knowledge that when a physical system deviates from the equilibrium state (for instance, when a semiconductor is placed in an external field) the fluctuation-dissipation theorem is violated. In this respect it is

pertinent to recall that both the Lifshitz theory combined with the Drude model and its modifications^{18–20} include transport phenomena in an external field and, thus, violate the applicability condition of the fluctuation-dissipation theorem on which they are based. The possibility of such violation is explicitly admitted by the statement¹⁸ that “It is not clear if the fields with the very low frequencies... are in thermal equilibrium with bodies. The problem is worth experimental investigation.” In our opinion experiments^{12–14} have already solved this problem in the most unambiguous manner.

6. Conclusions

From the foregoing we arrive to the following conclusions.

- 1) For metals with perfect crystal lattices the Lifshitz theory combined with the Drude model violates the Nernst theorem. The Nernst theorem is satisfied when the relaxation is nonzero at zero temperature, i.e. when impurities are taken into account. The Lifshitz theory including the dc conductivity of dielectrics and modifications of this theory violate the Nernst theorem for wide classes of different materials.
- 2) The experimental data of several experiments are inconsistent with the Lifshitz theory combined with the Drude model or including the dc conductivity and with the modifications of this theory. Keeping in mind that the Drude relation correctly describes the response of a metal to real (external) electric field, the reason of this inconsistency might be connected with some fundamental differences between real and fluctuating fields.
- 3) Phenomenologically, contradictions of the Lifshitz theory with both the Nernst theorem and the experimental data disappear if the free charge carriers are described by means of the plasma model in metals and are disregarded in dielectrics. Similar to any phenomenological approach, this one is useful as a practical matter but cannot be offered as an alternative to a complete theoretical description which remains unknown.
- 4) In our opinion, there are concepts of statistical physics related to the theoretical description of the interaction of classical and quantum fluctuating fields with matter that might need a reconsideration. Opinions on this subject vary and the consensus is not yet achieved.

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